

***IN-SITU* OXYGEN ISOTOPIC COMPOSITION OF TAGISH LAKE: AN  
UNGROUPED TYPE 2 CARBONACEOUS CHONDRITE**

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## Abstract

We have measured the oxygen isotopic composition of several components of Tagish Lake by ion microprobe. This meteorite constitutes the best preserved sample of C2 matter presently available for study. It presents two different lithologies (carbonate-poor and -rich) which have fairly comparable oxygen isotopic composition, with regard to both the primary or secondary minerals. For the olivine and pyroxene grains, their  $\delta^{18}\text{O}$  values range from - 10.5‰ to + 7.4‰ in the carbonate-poor lithology, with a mean  $\Delta^{17}\text{O}$  value of  $-3.7 \pm 2.4$ ‰. In the carbonate-rich lithology,  $\delta^{18}\text{O}$  varies from - 7.9‰ to + 3.3‰, and the mean  $\Delta^{17}\text{O}$  value is  $-4.7 \pm 1.4$ ‰. Olivine inclusions ( $\text{Fo}_{>99}$ ) with extreme  $^{16}\text{O}$ -enrichment were found in both lithologies :  $\delta^{18}\text{O} = -46.1$ ‰,  $\delta^{17}\text{O} = -48.3$ ‰ and  $\delta^{18}\text{O} = -40.6$ ‰,  $\delta^{17}\text{O} = -41.2$ ‰ in the carbonate-rich lithology;  $\delta^{18}\text{O} = -41.5$ ‰,  $\delta^{17}\text{O} = -43.4$ ‰ in the carbonate-poor lithology. Anhydrous minerals in the carbonate-poor lithology are slightly more  $^{16}\text{O}$ -rich than in the carbonate-rich one. Four low-iron manganese-rich (LIME) olivine grains do not have an oxygen isotopic composition distinct from the other "normal" olivines.

The phyllosilicate matrix presents the same range of oxygen isotopic compositions in both lithologies :  $\delta^{18}\text{O}$  from  $\sim 11$ ‰ to  $\sim 6$ ‰, with an average  $\Delta^{17}\text{O} \sim 0$ ‰. Because the bulk Tagish Lake oxygen isotopic composition given by Brown et al. (2000) is on the high end of our matrix analyses, we assume that this "bulk Tagish Lake" composition probably only represents that of the carbonate-rich lithology. Calcium carbonates have  $\delta^{18}\text{O}$  values up to 35‰, with  $\Delta^{17}\text{O} \sim 0.5$ ‰. Magnetite grains present very high  $\Delta^{17}\text{O}$  values  $\sim +3.4$ ‰  $\pm 1.2$ ‰.

Given our analytical uncertainties and our limited carbonate data, the matrix and the carbonate seem to have formed in isotopic equilibrium. In that case, their large isotopic fractionation would argue for a low temperature (CM-like,  $T \sim 0^\circ$ ) formation. Magnetite probably formed during a separate event. Tagish Lake magnetite data is surprisingly compatible

with that of R-chondrites (Greenwood et al., 2000) and unequilibrated ordinary (LL3) chondrites (Choi et al., 1998).

Our oxygen isotope data strongly supports the hypothesis of a single precursor for both lithologies. Drastic mineralogical changes between the two lithologies not being accompanied with isotopic fractionation seem compatible with the alteration model presented by Young et al. (1999).

Tagish Lake probably represents the first well preserved large sample of the C2 matter that dominates interplanetary matter since the formation of the solar system (see Gounelle et al., 2001a).

## 1. Introduction

The Tagish Lake meteorite is probably one of the best preserved and uncontaminated primitive carbonaceous chondrites available for study. It was collected frozen and in clean conditions soon after its fall (Brown et al., 2000). This meteorite is unique since its chemical and mineralogical compositions suggest similarities with both the CI1 and the CM2 carbonaceous chondrites. Tagish Lake presents two distinct lithologies ; the most abundant one is "carbonate-poor", and the relatively more minor lithology is "carbonate-rich" (e.g. Gounelle et al., 2001b). In terms of anhydrous components, Tagish Lake high temperature components bear more similarities with CM2 chondrites (especially with regard to the CAIs) (e.g. Gounelle et al., 2001b; Grossman and Simon, 2001; Simon and Grossman, 2001), although there are similarities also with CI (for instance the presence of MnO-rich olivines which are more common in CI1 than in CM2, see Klöck et al., 1989; and Simon and Grossman, 2001). The hydrous silicate component of Tagish Lake shows more similarities with the CI chondrites (Gounelle et al., 2001b; Grossman and Simon, 2001; Keller and Flynn, 2001; Mikouchi et al., 2001; Simon and Grossman, 2001). Although it was first proposed a type CI2 for this meteorite (Brown et al., 2000), thus meaning that Tagish Lake would be less altered than the other CI chondrites, there are still much discrepancy between Tagish Lake and a CI classification. It has been thus proposed that this meteorite represents a new type 2 carbonaceous chondrite (e.g. Gounelle et al., 2001b; Mikouchi et al., 2001).

It has been shown by analyses of whole-rock meteorites (Clayton, 1993; Clayton and Mayeda, 1999) and by recent ion microprobe studies, that the oxygen isotopic abundances vary on planetary to microscopic scales (e.g. Yurimoto et al., 1994; Leshin et al., 1997; Choi et al., 1998; Jones et al., 1998; McKeegan et al., 1998; Yurimoto et al., 1998; Engrand et al., 1999b; Hiyagon and Hashimoto, 1999; Greenwood et al., 2000; Leshin et al., 2000; Engrand et al., 2001; Leshin



et al., 2001). These variations provide one of the best records of the complex history of meteorites, asteroids and planetary bodies.

The oxygen isotopic composition of bulk fragments of Tagish Lake (Brown et al., 2000) shows differences with that of CI or CM chondrites. Recent ion microprobe analyses have allowed a comparison of the oxygen isotopic composition of Tagish Lake with other meteorites at the individual mineral level (Engrand et al., 2001; Leshin et al., 2001).

To understand better the history of Tagish Lake on its parent-asteroid, we report here a study of the oxygen isotopic composition of individual minerals measured by ion microprobe in both the carbonate-poor and the carbonate-rich lithology.

## **2. Samples and methods:**

Thick sections were made from small fragments of the meteorite. Both the carbonate-poor and the carbonate-rich lithology were analyzed (see companion paper by Zolensky et al., 2001, for a detailed description of the meteorite's mineralogy). In the carbonate-poor lithology, forty-five olivine, 3 pyroxene, 4 magnetite, and 1 small carbonate grain were analyzed. Eighteen spots on the matrix were also analyzed. In the carbonate-rich lithology, 13 olivine, 4 pyroxene, 2 calcium carbonate grains as well as 9 matrix locations were analyzed.

The textures and of the sample were analyzed by scanning electron microscope. The mineralogical composition was measured with a CAMECA CAMEBAX electron microprobe operated at 15 kV and 20 nA. Carbonates were analyzed at 15 kV and 4 nA. A focused beam ( $\sim 2\ \mu\text{m}$ ) was used for mineral analyses. Natural mineral standards were used, and corrections were applied for absorption, fluorescence, and atomic number effects using the CAMECA on-line PAP program. The oxygen isotopic composition was measured *in situ* with the Nancy CAMECA IMS1270 ion microprobe at high mass resolving power on an electron multiplier and

a faraday cup, following the procedure described in (Engrand et al., 1999b). Samples were coated with  $\sim 350 \text{ \AA}$  of gold. The  $\sim 0.4 \text{ nA}$  primary  $\text{Cs}^+$  beam was shaped into a spot of  $\sim 10 \times 20 \text{ }\mu\text{m}$ . Charge compensation was achieved using a normal-incidence electron flood gun (Slodzian, 1980).

Oxygen isotopic compositions are reported in permil as  $\delta^{17,18}\text{O}$  values calculated with regard to the SMOW value :

$$\delta^i\text{O} (\text{‰}) = \{ (^i\text{O}/^{16}\text{O})_{\text{sample}} / (^i\text{O}/^{16}\text{O})_{\text{SMOW}} - 1 \} \times 1000,$$

where  $i = 17$  or  $18$ ,  $(^{18}\text{O}/^{16}\text{O})_{\text{SMOW}} = 2.0052 \times 10^{-3}$  and  $(^{17}\text{O}/^{16}\text{O})_{\text{SMOW}} = 3.8288 \times 10^{-4}$ .

The  $\Delta^{17}\text{O}$  values will be also used, as they give a convenient representation of the deviation of the data from the terrestrial fractionation (TF) line :  $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$

Instrumental mass fractionation was corrected using a San Carlos olivine with  $\delta^{18}\text{O} = +5.25\text{‰}$  (Eiler et al., 1995), a magnetite standard ("Centre Afrique",  $\delta^{18}\text{O} = 1.42\text{‰}$ ), and a calcite standard ("Optical Calcite",  $\delta^{18}\text{O} = 16.12\text{‰}$ ). Olivine data were corrected for matrix effect at the level of  $+0.5\text{‰/amu}$  for an increase of  $\text{Fo}_{10}$  compared to the composition of the San Carlos olivine ( $\text{Fo}_{90.8}$ ). These corrections for olivine grains are usually at the level of  $1\text{‰}$ , and do not affect the interpretation of the data. Matrix effect corrections were not attempted for the analyses in the meteorite matrix, given its complex mineralogy. San Carlos olivine was taken as primary standard to correct for instrumental mass fractionation of the matrix analyses. This lack of a proper hydrous silicate standard could have introduced a systematic bias in the matrix analyses, whose extent is difficult to assess. However, this does not influence the discussion of the  $\Delta^{17}\text{O}$  matrix values, which are independent of a matrix correction which would only shift the data on a slope one-half (i.e. with constant  $\Delta^{17}\text{O}$ ) line. Furthermore, the matrices analyzed in both lithologies have a fairly comparable range of Mg numbers ( $\text{Mg}^*$  from  $\sim 40$  to  $\sim 80$ ). We therefore trust that it is legitimate to make a comparison between the  $\delta^{18}\text{O}$  and  $\delta^{17}\text{O}$  values measured in the matrices of both lithologies.

In our experimental conditions, the  $1\sigma$  uncertainty on the analysis of individual minerals is typically on the order of 1 - 1.5‰ for both  $\delta^{18}\text{O}$  and  $\delta^{17}\text{O}$  values. This reflects the internal measurement precision quadratically summed with the standard deviation of analyses on the primary standard during the analytical session (which represents the uncertainty in the instrumental mass fractionation correction and in the relative collection efficiency of the two detectors).

### 3. Results

#### 3.1. Texture and mineralogy

##### 3.1.1. Carbonate-poor lithology

The carbonate poor lithology is the dominant lithology (~ 80% in number of sections analyzed). It is characterized by a phyllosilicate-dominated matrix (mostly composed of fine-grained saponite including small sulfides and coarse grained saponite intergrown with serpentine and larger sulfide minerals, Mikouchi et al., 2001), containing rare chondrules, sparse CAIs, isolated olivines, magnetite and sulfides (Gounelle et al., 2001b and see Figure 1). Chondrules exhibit a thick (~200  $\mu\text{m}$ ) rim of phyllosilicates (Gounelle et al., 2001b; Simon and Grossman, 2001). Magnetite framboids are abundant, Ca-Mg-Fe carbonates are present but rare (Gounelle et al., 2001b; Zolensky et al., 2001).

We analyzed the oxygen isotopic composition of primary and secondary minerals from this lithology : pyroxene minerals have compositions  $\text{En}_{54.5-96.2}\text{Wo}_{1.2-2.8}$ , and olivine grains range in composition from  $\text{Fo}_{45}$  to  $\text{Fo}_{99.5}$ . Four olivine grains with a low-iron content and manganese-rich ("LIME" olivines), like those first reported by Klöck et al. (1989) were analyzed (Table 1, TL3-1-1, -2, -3, -26). They contain ~ 1.3 wt% MnO. One small Fe-Mg-Ca carbonate was also analyzed. It is however too small for a good quantitative analysis and the ion beam spot overlapped with some of the surrounding matrix. We analyzed framboidal magnetite inclusions

from this lithology, that are very pure in composition (less than 0.3 wt% of minor elements). The chemical composition of the spots analyzed in the matrix have Mg numbers which range from 34.3 to 78.4.

### 3.1.2. Carbonate-rich lithology

The rare carbonate-rich lithology (~ 20% in number) was described by several authors (e.g. Gounelle et al., 2001b; Mikouchi et al., 2001; see also Zolensky et al., 2001). The main differences with the carbonate-poor lithology (see Figure 2 for a micrograph of the carbonate-rich lithology) reside in the fact that CAIs are not present, there are no fine-grained clasts, the phyllosilicates is mainly saponite (Keller and Flynn, 2001), magnetite is rare, Fe-Mg-Ca carbonates are very abundant but calcium carbonates are very rare.

In this lithology, thirteen olivines with  $Fo_{91.3-99.2}$ , occurring both isolated in the matrix and within an aggregate have been analyzed. A radiant pyroxene chondrule was analyzed with 4 spots on the pyroxene minerals ( $En_{92-98}Wo_{1-4}$ ) and two spots on a mixture of pyroxene and interstitial mesostasis. Nine analyses were also made in the phyllosilicate-rich matrix. The two carbonates analyzed are almost pure calcium carbonates containing no detectable Mg or Mn and having a FeO content below 0.34 wt%. The carbonate population is dominated by fine-grained breunnerite (Gounelle et al., 2001b), but they are too small to be analyzed free of the surrounding matrix. Thus although calcium carbonates are rare in this lithology, they were analyzed preferentially here because of their larger sizes (~ 20  $\mu m$ ). For this lithology, the Mg numbers for the spots analyzed in the matrix are fairly comparable to those of the carbonate-poor lithology; they range from 42.7 to 83.0.



### 3.2. Oxygen isotopic composition:

As will be detailed in the discussion, the oxygen isotopic compositions of the carbonate-rich and the carbonate-poor lithologies are fairly comparable. For clarity purpose, the results for each lithology are presented separately in the tables (see Table 1 and 2). Data for all minerals are represented for each lithologies in Figures 3 and 4, and Figures 5 and 6 show data for primary and secondary minerals, respectively, for both lithologies.

#### 3.2.1. Primary minerals : olivines and pyroxenes

##### 3.2.1.1. Carbonate-poor lithology

The data fall between the Young & Russell line (hereafter abbreviated as Y&R line, see (Young and Russell, 1998) and the CAI line (Clayton, 1993). For the olivine grains, the oxygen isotopic composition range in  $\delta^{18}\text{O}$  from  $-10.5\text{‰}$  to  $+7.4\text{‰}$ , with  $\delta^{17}\text{O}$  values from  $-14.1\text{‰}$  to  $+5.4\text{‰}$  (Table 1a, Figures 3 and 5). The four LIME olivine do not have oxygen isotopic compositions distinct from the other olivines (Table 1a).

Two olivine inclusions ( $\text{Fo}_{>99}$ ) with extreme  $^{16}\text{O}$ -enrichment were found ( $\delta^{18}\text{O} = -46.1\text{‰}$  and  $\delta^{17}\text{O} = -48.3\text{‰}$ ;  $\delta^{18}\text{O} = -40.6\text{‰}$  and  $\delta^{17}\text{O} = -41.2\text{‰}$ ) (Table 1a, Figures 3a and 7).

For the pyroxene minerals, the  $\delta^{18}\text{O}$  values vary from  $-10.0\text{‰}$  to  $+8.8\text{‰}$ , and  $\delta^{17}\text{O} = -11.8\text{‰}$  to  $+5.0\text{‰}$  (Table 1a, Figures 3 and 5).

The average oxygen isotopic composition for the anhydrous minerals (except for the extreme  $^{16}\text{O}$ -rich olivines) is  $\delta^{18}\text{O} = -3.0\text{‰}$  and  $\delta^{17}\text{O} = -5.3\text{‰}$ . The corresponding  $\Delta^{17}\text{O}$  values are typically in the range observed for carbonaceous chondrites, between  $\sim -8\text{‰}$  and  $0\text{‰}$ , with an average value at  $-3.7 \pm 2.4\text{‰}$  ( $1\sigma$ ).

There is no simple correlation between the chemical composition of the olivine and their oxygen isotopic composition. However, the general pattern (see Figure 8) observed in



carbonaceous chondrite and in Antarctic micrometeorite olivine grains (Leshin et al., 1997; Engrand et al., 1999b; Leshin et al., 2000) is also found in Tagish Lake.

#### 3.2.1.2. Carbonate-rich lithology

The data fall roughly in the same range as for the carbonate poor lithology. Most of the olivine grains show a "normal" isotopic composition ( $\delta^{18}\text{O} = -7.9\text{‰}$  to  $+3.3\text{‰}$ , with  $\delta^{17}\text{O}$  from  $-11.1\text{‰}$  to  $-0.1\text{‰}$ ; Table 2, Figures 4 and 5). However, two olivine grains in an aggregate exhibit very  $^{16}\text{O}$  enriched values :  $\delta^{18}\text{O} = -40.6\text{‰}$  and  $-42.5\text{‰}$ , with  $\delta^{17}\text{O} = -43.4\text{‰}$  and  $-43.3\text{‰}$ , respectively. This corresponds to the same degree of enrichment described by Hiyagon and Hashimoto (1999) and Yurimoto and Wasson (2000) in the meteorites Yamato-86009 (CV3), Murchison (CM2) and Y81020 (CO3).

Figure 8 shows the pattern observed for the oxygen isotopic composition of the olivine as a function of their iron content. This pattern is comparable in both lithologies and is compatible with that previously observed in carbonaceous chondrites (Leshin et al., 1997; Leshin et al., 2000) and in Antarctic micrometeorites (Engrand et al., 1999b).

The oxygen isotopic composition of a radiant pyroxene chondrule has a bimodal distribution (Table 2 and Figure 4b). The pyroxene minerals present  $\delta^{18}\text{O}$  values between  $-8.7\text{‰}$  and  $-6.2\text{‰}$ , with  $\delta^{17}\text{O}$  from  $-9.4\text{‰}$  and  $-7.3\text{‰}$ , respectively. In the analysis spots which overlap on the mesostasis,  $\delta^{18}\text{O}$  values vary between  $+6.4\text{‰}$  and  $+8.4\text{‰}$ , with  $\delta^{17}\text{O}$  from  $+2.1\text{‰}$  to  $+2.7\text{‰}$ .

The average isotopic composition for the "normal" (i.e. non extreme  $^{16}\text{O}$ -rich) anhydrous minerals is comparable with those of the carbonate-poor lithology :  $\delta^{18}\text{O} = -2.8\text{‰}$  and  $\delta^{17}\text{O} = -5.7\text{‰}$ . The  $\Delta^{17}\text{O}$  values for these olivine and pyroxene grains give an average value of  $-4.7 \pm 1.4\text{‰}$  ( $1\sigma$ ).

### 3.2.2. Secondary minerals : matrix, carbonates and magnetite

#### 3.2.2.1. Carbonate-poor lithology

The oxygen isotopic composition of the matrix in the carbonate-poor lithology spreads roughly along the terrestrial fractionation line (see Table 1b, Figures 3 and 6), with  $\delta^{18}\text{O}$  from +3.6‰ to +19.7‰ and  $\Delta^{17}\text{O}$  from -1.8‰ to +3.1‰ (with an average  $\delta^{18}\text{O}$  = +11.0‰ and average  $\Delta^{17}\text{O}$  value of +0.5‰).

Carbonates in this lithology are rare and small. Data for only one carbonate was obtained :  $\delta^{18}\text{O}$  = +25.5‰, with  $\delta^{17}\text{O}$  = +14.0‰ (Table 1b, Figures 3 and 6). This value correspond to  $\Delta^{17}\text{O}$  = +0.7‰.

Data for the magnetite grains are significantly above the terrestrial fractionation (TF) line. Their  $\delta^{18}\text{O}$  values range from -6.2‰ to -9.0‰, with  $\Delta^{17}\text{O}$  = +2.5‰ to +4.4‰ (Table 1b and Figure 6). The "magnetite" data points found closer to the terrestrial fractionation line are interpreted as the result of physical mixing between magnetite and matrix in the ion beam spot, since the framboidal magnetite grains can be very small (the data points are labeled "Mt + Mx", for "magnetite + matrix").

#### 3.2.2.2. Carbonate-rich lithology

The hydrous silicate data as well as the carbonate data spread about on the TF line above its intercept with the CAI line (see Figures 4 and 6). The  $\delta^{18}\text{O}$  values vary from +1.4‰ to +20.3‰, with  $\Delta^{17}\text{O}$  values from -1.8‰ to +2.9‰ (average values :  $\delta^{18}\text{O}$  = +11.3‰;  $\Delta^{17}\text{O}$  = 0.0‰). These matrix analyses are compatible with the bulk analyses reported in (Brown et al., 2000) and with CI or CM matrix analyses (Clayton and Mayeda, 1984; Mayeda et al., 1987; Rowe et al., 1994; Clayton and Mayeda, 1999);

Two calcium carbonate grains show heavy oxygen isotopes enrichment :  $\delta^{18}\text{O} = +25.0\text{‰}$  and  $+35.3\text{‰}$ , with  $\delta^{17}\text{O} = +13.7\text{‰}$  and  $+18.8\text{‰}$ , respectively. These values are compatible with the analyses of Leshin *et al.* (2001 ) and (Grady *et al.*, 2001). They are also roughly compatible with what is usually observed in carbonaceous chondrite carbonates (e.g. Grady *et al.*, 1988; Zito *et al.*, 1998).

## **4. Discussion:**

### **4.1. The Tagish Lake bulk oxygen isotopic composition revisited**

Mineralogical study of the Tagish Lake meteorite demonstrates that it is a breccia (see Zolensky *et al.*, 2001, and above), mainly made of a major lithology (the carbonate-poor), and of a minor lithology (the carbonate-rich). During the preliminary examination phase of Tagish Lake (i.e. Brown *et al.*, 2000), the existence of two main lithologies had not been recognized yet. The presence of the carbonate-rich lithology was first acknowledged at the 2001 Houston conference (Gounelle *et al.*, 2001b).

Since the highly brecciated nature of Tagish Lake was yet unknown, no special care has been taken to characterize the mineralogy of the fragments used for measuring the meteorite bulk oxygen isotopic composition with a fluorination technique (Brown *et al.*, 2000; Clayton and Mayeda, 2001). The two bulk fragments measured lead to somewhat similar oxygen isotopic compositions :  $\delta^{18}\text{O} = +19.0\text{‰}$  and  $+18.0\text{‰}$ ,  $\delta^{17}\text{O} = +9.2\text{‰}$  and  $+8.3\text{‰}$ , respectively (Brown *et al.*, 2000; Clayton and Mayeda, 2001). On a three isotope plot (see Figure 3 and 4, or Figure 6), it is shown that the bulk Tagish Lake oxygen isotopic composition measured by the fluorination technique is heavier than most of the mineral phases (i.e. anhydrous silicates, matrix, magnetite) measured with the ion microprobe, but carbonates (and marginally in the matrix). Since Tagish Lake is a mixture of individual phases, its heavy bulk oxygen isotopic composition means that carbonates are probably a major phase of the fragment measured by the fluorination technique

(Brown et al., 2000; Clayton and Mayeda, 2001). Because carbonates are abundant only in the carbonate-rich lithology, we suggest that the so-called bulk meteorite oxygen isotopic composition is most probably the bulk oxygen isotopic composition of the carbonate-rich lithology, the relatively minor one.

The anhydrous minerals and the matrix oxygen isotopic composition are fairly comparable in the two lithologies (see Figures 5 and 6). Because the fine-grained matrix and the anhydrous minerals are the two main phases of the carbonate-poor lithology and since carbonates will contribute only little to its oxygen isotopic composition, we predict that the bulk oxygen composition of the carbonate-poor lithology should be lighter than that of its carbonate-rich counterpart. The carbonate-poor lithology being the major one, we suggest that the bulk oxygen isotopic composition of the Tagish Lake meteorite is probably lighter than the composition published by Brown *et al.* (2000) and Clayton and Mayeda (2001).

Although other anomalous C2 chondrites have an oxygen isotopic composition in the range of CM2 meteorites (Clayton and Mayeda, 1999), this is not the case for Tagish Lake. However, if the bulk oxygen isotopic composition of Tagish Lake is lighter than thought (see above), it brings this unusual meteorite closer to the range found for CM2 chondrites on an oxygen three isotope diagram. Our conclusions regarding the bulk oxygen isotopic composition of Tagish Lake strengthen the links with CM2 meteorites and weaken the link CI1 meteorites. We consider that the relationship of Tagish Lake with C2 meteorites is stronger than with any other group of carbonaceous chondrites.

#### **4.2. Primary mineral formation (pyroxene and olivine grains)**

The oxygen isotopic compositions measured in primary minerals from both lithologies are rather comparable (Figure 5). The olivine and the pyroxene grains in the carbonate-poor lithology span a slightly larger range of in the three isotope diagram, the extreme compositions



for both minerals in the carbonate-poor lithology being at the same time slightly more  $^{16}\text{O}$ -rich and slightly more  $^{16}\text{O}$ -poor than their counterparts in the carbonate-rich lithology (Figure 5). We cannot exclude however that this might not be a sampling effect, as more data were taken from the carbonate-poor lithology.

The oxygen isotopic composition measured in the pyroxene chondrule shows at the same time very primitive values plotting on the Young and Russell (Y&R) line (Young and Russell, 1998) and values which are very close to that of some part of the matrix (see Figure 4, and see discussion in the next section).

Our olivine and pyroxene data is in general compatible with previous data measured on individual minerals in carbonaceous chondrite (Leshin et al., 1997; Leshin et al., 2000; Leshin et al., 2001). The observed slight  $^{16}\text{O}$ -enrichment observed here in the "normal" olivine grains in the carbonate-poor lithology is still not as high as that found by Leshin *et al.* (2001). The range of variation of oxygen isotopic compositions measured in individual minerals in carbonaceous chondrites does not usually allow to distinguish between the different classes of carbonaceous chondrites (Leshin et al., 1997; Leshin et al., 2001). This is also the case here, where our data on anhydrous minerals of Tagish Lake could be compatible with all classes of carbonaceous chondrites. Our present data are also comparable with the oxygen isotopic composition of Antarctic micrometeorite individual minerals (see Engrand et al., 1999b).

We found in Tagish Lake four low iron and manganese-rich olivine (LIME) grains. These olivine whose Mn content ranges up to 1.3 wt % MnO are rare objects known to be present in Stratospheric Interplanetary Dust Particles (SIDPs), unequilibrated ordinary chondrites (UOC) (see Klöck et al., 1989 for a first description of these objects) and Antarctic micrometeorites (Devouard and Gounelle, 2001 unpublished data). Klöck *et al.* (1989), who first identified these



puzzling grains, proposed that these olivine originated by condensation from a gas of solar composition. We have for the first time been able to measure the oxygen isotopic composition of LIME olivines, and found that they have a "normal" (i.e., non  $^{16}\text{O}$ -enriched) composition (see Table 1b). This finding could be compatible with the origin proposed by Klöck *et al.* (1989), since some olivine-rich chondrules are supposed to have formed by condensation (i.e. Krot *et al.*, 2001), and most chondrules do not exhibit any significant  $^{16}\text{O}$  enrichment (e.g. Leshin *et al.*, 2000). On the other hand, their oxygen isotopic composition cannot be used to argue for an origin by condensation. The fact that these LIME olivine have been found in the most primitive objects (SIDPs, Antarctic micrometeorites...) supports the primitivity of Tagish Lake.

In addition, extreme  $^{16}\text{O}$ -rich olivine grains were found in both lithologies, and the above mentioned trend of a higher  $^{16}\text{O}$  enrichment in the carbonate-poor lithology compared to the other lithology also holds true for the extreme  $^{16}\text{O}$ -rich olivine aggregates (Table 1b and 2, and Figure 3 and 4). This abundance of extreme  $^{16}\text{O}$ -rich olivines in this meteorite seems rather high, since 3 such inclusions have been found in a few millimeter-sized fragments.

The discovery of very  $^{16}\text{O}$  enriched olivines in Tagish Lake like what was found by Hiyagon and Hashimoto (1999) and Yurimoto and Wasson. (2000) supports the very primitive nature of this meteorite. Clayton and Mayeda (1984; 1999) propose that the "normal" oxygen isotopic composition of olivine and pyroxene grains usually found in carbonaceous chondrites (i.e. those with no extreme  $^{16}\text{O}$  enrichment) formed by interaction of anhydrous mineral precursors with  $\delta^{18}\text{O} \sim \delta^{17}\text{O} \sim -40\text{‰}$  and a nebular gas rich in heavy oxygen isotopes. In the frame of this model, the carbonate-poor lithology being a little more  $^{16}\text{O}$ -rich than the carbonate-poor one is probably the most primitive of the two, as is also suggested by the mineralogy (see above and Zolensky *et al.*, 2001).

This extreme  $^{16}\text{O}$  enrichment suggests a common oxygen source or a common chemical process for both CAIs and this kind of olivine. It is striking that so far such an enrichment in olivine grains has only been observed in primitive carbonaceous chondrites although  $^{16}\text{O}$  enrichments have been observed in CAI minerals from carbonaceous chondrites as well as in ordinary chondrites CAIs (McKeegan et al., 1998).

As a whole, the oxygen isotopic compositions measured for anhydrous minerals from both lithologies are fairly comparable (Figure 5). This observation strongly suggests that both lithologies are issued from a unique precursor, and suffered different alteration history. In fact, the mineralogical observations agree with this conclusion (Zolensky et al., 2001). In the next section, we will try to decipher this alteration history from the study of the oxygen isotopic composition of secondary minerals.

#### **4.3. Alteration history recorded by the oxygen isotopic composition of secondary minerals**

Tagish Lake is a fantastic meteorite that enables planetary scientists to scout the progression of aqueous alteration in a primitive solid body in some unprecedented way. Tagish Lake hosts two main lithologies that are mineralogically quite distinct, with regard to the abundance and the composition of secondary minerals (mesostasis, phyllosilicates, carbonates, magnetite, sulfides, etc...). Careful mineralogical examination of the meteorite (this work and Zolensky et al., 2001), as well as the oxygen isotopic of the anhydrous minerals being slightly more  $^{16}\text{O}$ -rich in the carbonate-poor lithology than in the carbonate-rich one, suggest that the carbonate-rich lithology has formed from the carbonate-poor lithology after some extra aqueous

alteration. The oxygen isotopic composition might help precise this genetic link between the two lithologies to track down the evolution during aqueous alteration.

We will first examine the differences between the oxygen isotopic compositions of secondary minerals from the two lithologies. Then we will compare the oxygen isotopic composition of secondary minerals in Tagish Lake and in CI1 and CM2 chondrites.

The oxygen isotopic composition of the matrices from the two lithologies look broadly similar (Figure 6). Within uncertainty, analyses of the matrix in the two lithologies lead similar average oxygen isotopic composition :  $\delta^{18}\text{O} = +11.3\text{‰}$  and  $+11.0\text{‰}$  ;  $\Delta^{17}\text{O} = +0.5\text{‰}$  and  $0.0\text{‰}$  for the carbonate-poor and -rich lithologies, respectively (Table 1b and 2 ; see also Figure 6). Since the matrix analyses have been performed with a  $\sim (10 \times 20) \mu\text{m}^2$  beam, they include a mixture of oxygen-bearing phases. In the carbonate-poor lithology, the main oxygen-bearing phase is the phyllosilicates, whereas in the carbonate-rich lithology, the main oxygen-bearing phase are the phyllosilicates, the magnetites and the carbonates. Since the carbonates in Tagish Lake have a very heavy oxygen isotopic composition (this work and Leshin et al., 2001), the phyllosilicates (saponite) from the carbonate-rich lithology may have a lighter composition than the phyllosilicates (saponite + serpentine) in the carbonate-poor lithology.

A radiant pyroxene chondrule from the carbonate-rich lithology exhibits a mesostasis with an oxygen isotopic composition close to that of the matrix (see Figure 4). Metzler *et al.* (1992) propose that, in the case of CM2 chondrites, the formation, fragmentation, alteration and rimming of the chondrules predate their incorporation in the parent body. Simon *et al.* (2001) suggest that if this is also the case for Tagish lake, then the silicate precursors were readily more altered than in CM2 chondrites before their incorporation in the parent body. However, the oxygen isotopic composition of the chondrule mesostasis in Tagish Lake being in the range of values measured for the matrix possibly indicates a contemporaneous interaction of the fluid

with the meteorite to form the phyllosilicates and with the interstitial glass in the chondrule. That would suggest that at least some chondrules were incorporated unaltered in the meteorite and that a later stage of alteration occurred in the parent body.

Only three carbonates could be analyzed in our Tagish Lake sample, thus our comparison has to be taken with caution. A sufficient amount of data on carbonates is missing to establish a pertinent comparison between the two lithologies. Although a fair amount of data is presented by Leshin *et al.* (2001), the lithology they studied is unknown. We suspect that the ~ 300 mg samples they studied contained both lithologies since they report an intermediate amount of carbonates. In such a case, as scarcely suggested by our data, no significant differences can be observed between the carbonate oxygen isotopic compositions in the two lithologies.

To summarize, only a hypothetical lighter oxygen isotopic composition of the carbonate-rich phyllosilicates is noted when comparing the oxygen isotopic composition of secondary minerals from the two lithologies.

*How does the oxygen isotopic composition of the C2 "ungrouped" Tagish Lake meteorite secondary minerals compare to the CM2 and CI1 analyses ?*

Six other ungrouped C2 chondrites from Antarctica (e.g. Clayton and Mayeda, 1999) have whole-rock data in the range of CM2 chondrites. An ungrouped C1 chondrite (Yamato 82162, Clayton and Mayeda, 1999) has an oxygen isotopic composition close to the CI field on the three isotope diagram. Some Tagish Lake matrix analyses are rather compatible with either CI1 or CM2 of analyses (see Figure 9). However, the other analyses present a higher  $\Delta^{17}\text{O}$  value than either CI1 or CM2 matrix.



The average  $\Delta^{17}\text{O}$  value of Tagish Lake matrix (0.0‰ to +0.5‰) is closer to the  $\Delta^{17}\text{O}$  range of the CI1 matrix analyses, whereas its range in  $\delta^{18}\text{O}$  values spreads over the whole range of  $\delta^{18}\text{O}$  values in CI1 and CM2 matrix analyses (see Figure 9).

The only reasonable conclusion we can draw so far, is that the oxygen isotopic composition of Tagish Lake matrix is intermediate between the CI1 and the CM2 matrix oxygen isotopic compositions.

Tagish Lake carbonate oxygen isotopic composition are broadly compatible with that of CI1s and CM2s ones (see Figure 10 ; see Grady et al., 2001 for  $\delta^{18}\text{O}$  values of Tagish Lake carbonates ; see data for Tagish Lake and Orgueil carbonates in Leshin et al., 2001), (see Benedix et al., 2000 for data on CM2 carbonates). Leshin et al. (2001) report that carbonates in Tagish Lake have the highest  $\Delta^{17}\text{O}$  ever reported in any carbonates. Given our large analytical uncertainties, the  $\Delta^{17}\text{O}$  values we measured are indistinguishable from that of the matrix.

Grady et al. (2001) observe that the C and O isotopic composition of Tagish Lake carbonates plot on the extreme end of the trend defined by Zito et al. (1998), suggesting that Tagish Lake might contain more calcium carbonates with regard to Mg-Fe carbonates than CI chondrites. However, an average value of  $\delta^{18}\text{O} \sim 32\text{‰}$  is found for calcium carbonates in Tagish Lake (Grady et al., 2001; Leshin et al., 2001). This value is significantly lower than the average  $\delta^{18}\text{O}$  calcite value ( $\sim 39\text{‰}$ ) measured in Orgueil by Zito et al. (1998), but is slightly higher than that of the calcite-rich fraction measured in Orgueil by Leshin et al. (2001). Zito et al. (1998) analyzed individual and well characterized calcite grains on a polished section. Our hypothesis is that the apparent discrepancy of  $\sim 7\text{‰}$  in  $\delta^{18}\text{O}$  in the analyses of calcite in Orgueil observed between the work of Zito et al. (1998) and that of Leshin et al. (2001) is due to the presence of other carbonates or impurities in the calcite-rich fraction identified from the release temperature in the latter case. We then consider that the true  $\delta^{18}\text{O}$  value for calcite in Orgueil is  $\sim 39\text{‰}$ , as



deduced from the work of Zito et al. (1998). The oxygen isotopic composition of the calcite we analyzed in Tagish Lake is then significantly lighter than that of Orgueil calcite grains.

Our data (given the associated analytical uncertainty) suggests that carbonates and matrix might have formed in isotopic equilibrium (see Figure 6). If this is the case, the large ( $\sim 24\text{‰}$ ) fractionation observed between the average  $\delta^{18}\text{O}$  value measured in the matrix ( $\sim 11\text{‰}$  for both lithologies) and the average  $\delta^{18}\text{O}$  value of the carbonates ( $\sim 35\text{‰}$ ; our data point and Grady et al. 2001) argues in favor of a CM-like temperature of  $\sim 0^\circ\text{C}$  (the fractionation factors for hydrous silicate - water and carbonate - water as a function of temperature are taken from O'Neil et al., 1969; and Zheng, 1993).

The average  $\Delta^{17}\text{O}$  we have measured for the matrix and the carbonates ( $\sim 0\text{‰}$ ) is compatible with that of the low temperature water measured by Baker et al. (2001). This low temperature component is observed by Baker et al. (2001) up to temperatures of  $\sim 400^\circ\text{C}$ , which is at the lower limit of the range of dehydration temperature of phyllosilicates.

Leshin et al. (2001), argue that the high  $\Delta^{17}\text{O}$  they measured for the carbonates in Tagish Lake corresponds to a altering water with high initial  $\delta^{18}\text{O}$  value, the corresponding isotopic fractionation observed on the carbonates being compatible with CI-like temperatures on the range ( $\sim 50 - 100^\circ\text{C}$ ). As the Tagish Lake whole-rock data falls on the mixing line previously observed for CO3 and CM2 chondrites (Clayton and Mayeda, 1999; Clayton and Mayeda, 2001), Clayton and Mayeda (2001) favor a low temperature (CM-like,  $\sim 0^\circ\text{C}$ ) for the formation of the carbonates in Tagish Lake. This model implies that, like in Murchison, the carbonates and the phyllosilicates formed in isotopic equilibrium (Clayton and Mayeda, 1984). Clayton and Mayeda (2001), in the frame of the closed system hydration model (Clayton and Mayeda, 1999), predict a very high water : rock ratio of 1.2 to reproduce the observed bulk oxygen isotopic composition of Tagish Lake. This value might however be lowered if the "true" bulk composition of Tagish Lake is lighter than the values adopted by Brown et al. (2000) (see above).

Flynn *et al.* (2001) observe the presence of a moderately volatile organic phase in the meteorite which suggest that Tagish Lake was not heated at a temperature higher than 100°C. But the epoch at which this moderately volatile component cannot be determined by comparison with the carbonate-forming event.

Confirmation of the formation condition for carbonates will require a dedicated study of the carbon and oxygen isotopic composition of Tagish Lake carbonates, and in particular of the breunnerite which is the most abundant carbonate in the Tagish Lake carbonate-rich lithology (Gounelle *et al.*, 2001b). This could not be done here in particular because of the very small size of the breunnerite grains.

Magnetites in Tagish Lake are in isotopic disequilibrium with carbonates and matrix, as their measured  $\Delta^{17}\text{O}$  values are significantly distinct ; they probably formed during a separate events, at different times or temperatures, from that which formed the matrix and the carbonates.

Magnetites from Tagish Lake have higher  $\Delta^{17}\text{O}$  values ( $\sim + 3.4\text{‰} \pm 1.2\text{‰}$  in average) than magnetites in CI1 chondrites :  $\Delta^{17}\text{O}_{\text{mt}} = + 1.7\text{‰}$  (average of 6 analyses from Rowe *et al.*, 1994) or in the CM2 chondrite Essebi where  $\Delta^{17}\text{O}_{\text{mt}} = + 1.3\text{‰}$  (Rowe *et al.*, 1994) (see Figure 11). The  $\delta^{18}\text{O}$  values of the Tagish Lake magnetite grains are lower than that of CI1 or CM2 chondrites (note that Essebi, Haripura and Bells are the only CM2 chondrites to have a high magnetite abundance).

Baker *et al.* (2001) report a high temperature component of the water in Tagish Lake which presents a  $\Delta^{17}\text{O}$  value of  $\sim 1\text{‰}$ . This probably represents a later stage of alteration, suggesting that magnetites formed in an earlier event from a water with higher  $\Delta^{17}\text{O}$ . Tagish Lake alteration

stopped far from equilibrium, as the difference between  $\Delta^{17}\text{O}$  of water and silicates ( $\sim 2\text{‰}$ ) is larger than in any other meteorite.

These values for magnetite are the highest ever measured in carbonaceous chondrite magnetites, and are surprisingly more compatible with those measured in R-chondrites ( $\text{D17O} = +2.5\text{‰}$  to  $+4.3\text{‰}$ , Greenwood et al., 2000) and marginally with those measured in unequilibrated ordinary chondrites (LL3) ( $\text{D17O} = +4$  to  $+7\text{‰}$ , Choi et al., 1998) (see Figure 11 and Fig. 8 in Greenwood et al., 2000).

Although our data is limited, we observe a narrow range in  $\delta^{18}\text{O}$  values of magnetites ( $\sim 3\text{‰}$ ). On the contrary to what is found in LL3 chondrites (Choi et al., 1998) and to a lower extent in R-chondrites (Greenwood et al., 2000), formation of Tagish Lake magnetites did not form from a limited water reservoir.

Choi et al. (1998) suggested that the  $\Delta^{17}\text{O}$  of the nebular gas increased with time. If that hold true, the Tagish Lake magnetites should have formed later than CM or CI magnetites. Data obtained by Busfield *et al.* (2001) also suggests that the formation of Tagish Lake magnetite was later than those in Orgueil by  $\sim 50$  Ma. The present data also indicates that Tagish Lake magnetite did not occur by replacement of olivine, like what is observed for magnetites in CV chondrites (Choi et al., 2000). They possibly formed by the same process of aqueous alteration of metal at the same time when magnetites in R chondrites and LL3 chondrites formed, probably on their respective parent body.

To summarize, the oxygen-bearing secondary minerals from Tagish Lake present a specific signature, different either from CM2 chondrites, or from CI1 chondrites. From our analyses we can deduce that there were probably two separate aqueous alteration events, as the ranges of  $\Delta^{17}\text{O}$  values measured for different groups of secondary phases (matrix and carbonates ;

magnetites) are distinct (although the uncertainty on our analyses is a little too large to make of it a very strong point).

Unfortunately, our carbonate data does not allow us a suitable comparison between the oxygen isotopic composition of carbonates from both lithologies. If we assume that there is no significant differences between the carbonate isotopic composition of carbonates between the two lithologies, we can propose the following sequence of alteration : phyllosilicates and carbonates form at low temperature ( $\sim 0^{\circ}\text{C}$ ) in isotopic equilibrium in the carbonate-poor lithology. Further alteration of part of this lithology (at the same time?) leads to the relatively minor carbonate-rich lithology without significant isotopic changes in the matrix, and (?) in the carbonates. The magnetites form later, or from a different fluid.

We now have to answer a question provoked by the previous examination of data : is the specific oxygen isotopic composition of Tagish Lake better explained by a model where aqueous alteration is specific to every parent-body/chondrite group, or by a model where dynamic aqueous alteration happens in a single parent-body enclosing all the carbonaceous chondrites groups ?

The important mineralogical change with no correlated changes in the oxygen isotopes might be explained by the aqueous alteration model proposed by Young *et al.* (1999). In this model, the initial amount of ice present is limited, and alteration occurs through the *dynamic flow* of melted ice. After 470 000 years of alteration, the fictive parent is radially zoned, with each zone corresponding to a different oxygen isotopic composition, and a different amount of secondary minerals. Each isotopic-mineralogical zone has its counterparts in the different chondritic groups. This zoning, as well as the correspondence with the actual chondritic groups, is however only qualitative, and because the alteration is dynamic -the oxygen isotopic



composition being controlled either by the rock, or by the water- it is possible that the strict correlation between oxygen isotopic composition and the mineralogy be blurred in some transition region. In the closed alteration model presented by Clayton and Mayeda (1999), the water is stagnant and very abundant. It seems then more difficult to juxtapose two such lithologies with both different mineralogical features and fairly similar oxygen isotopic compositions.

The versatility of the model proposed by Young et al. (1999) makes it easy to incorporate the Tagish Lake oxygen isotopic fingerprint. The matrix and the carbonate data reported here overlap the range of oxygen isotopic composition reported by in the Figure 3 of Young *et al.* (1999). However, we would like to question the fact that the Young model takes into consideration an alteration episode lasting only 470 000 years, when the I-Xe systematics suggest that alteration in Tagish Lake have lasted as long as 50 Ma (Busfield et al., 2001).

#### **4.4. The predominance of C2 matter in interplanetary space since the formation of the solar system.**

It is argued that Tagish Lake could represent the micrometeorite's long-sought-for parent body. In particular, Noguchi et al. (2001) were able to reproduced the textures and composition of phyllosilicates, as well as the frequent occurrence of magnesiowüstite observed in Antarctic micrometeorites by heating fragments of Tagish Lake in an attempt to reproduce the atmospheric entry heating.

Based on a TEM study, Keller and Flynn (2001) also suggest that Tagish Lake matrix mineralogy is similar to that of hydrated SIDPs.



However, the D/H ratios measured in Tagish Lake (Messenger, 2001; Pearson et al., 2001) show moderate enrichments in deuterium which are not present in Antarctic micrometeorites (Engrand et al., 1999a).

Based on reflectance spectroscopy measurements, Hiroi et al. (2001) proposed in turn a source for Tagish Lake from a D-type asteroid.

The fact that Tagish Lake might indeed be the micrometeorites' parent-body association will still have to be demonstrated, but it is a fact that both Tagish Lake and the micrometeorites are constituted of an anomalous C2 matter which seems to constitute the extraterrestrial matter dominating the interplanetary space. Furthermore, there are now evidences that C2 matter has dominated the interplanetary matter over the history of solar system (see Gounelle et al., 2001a).

In the model proposed by Young *et al.* (1999) the C2 matter is present in the middle of the fictive parent-body. There is no physical reason to retrieve more of C2 matter than the type 3 material that is on the outside of the body. One hypothesis could consider that it takes  $\sim 4.5$  billion years to peel off the C3 primitive crust of the asteroid to reveal the C2 matter, but this argument does not hold if C2 matter has dominated the interplanetary matter since the formation of the solar system (e.g. Gounelle et al., 2001a). There are actually also some evidences for C2 meteorites being regolith breccias, which would be objects contradictory with the C2 being inside rather than outside a fictive parent-body.

The origin of Tagish Lake (and of micrometeorites) will still have to be debated, but it seems more and more unavoidable that such a C2 matter is surrounding us from everywhere and that only a small fraction of it survives the atmospheric entry and weathering in the terrestrial environment, thus explaining the apparent overwhelming abundance of tougher ordinary chondrites in the meteorite collections.

## 5. Conclusions

Tagish Lake is a unique meteorite recovered in unprecedented clean conditions. It presents two different lithologies (carbonate-poor and -rich) with very distinct mineralogical features which are intermediate between C1 and C2 characteristics. A detailed study speaks in favor of a C2 classification, although the meteorite is different from other known C2 meteorites (e.g. Gounelle et al., 2001b; Zolensky et al., 2001).

The oxygen isotopic compositions of both lithologies are fairly comparable, with regard to both the primary or secondary minerals. Anhydrous minerals in the carbonate-poor lithology are slightly more  $^{16}\text{O}$  enriched than in the carbonate-rich one. This strongly supports the hypothesis of a single precursor for both lithologies.

The oxygen isotopic composition of four LIME olivine is not significantly distinct from that of the other olivines.

As the oxygen isotopic composition measured on a bulk Tagish Lake sample is on the upper limit of the range found for our matrix analyses, we suggest that the Tagish Lake bulk composition considered by Brown et al. (2000) represents that of the carbonate-rich lithology. The "true" bulk oxygen isotopic composition might then plot closer to the C2 field. The oxygen isotopic compositions of Tagish Lake secondary minerals plots closer to that of C2 matter than to CI1 material, but are still distinct from both.

Although our data is (very) limited for the carbonates, the phyllosilicates (matrix) and the carbonates seem to have formed in isotopic equilibrium, with no difference between the oxygen isotopic composition of these secondary minerals in both lithologies. Magnetite is present only in the carbonate-poor lithology and seems to have formed during a separate event. The  $\Delta^{17}\text{O}$  values found for the magnetites are higher than for any other carbonaceous chondrites. Surprisingly, they are compatible with magnetite data from R-chondrites (Greenwood et al., 2000) and unequilibrated ordinary (LL3) chondrites (Choi et al., 1998).

Our data where drastic mineralogical changes do not seem accompanied with different isotopic fractionation seem compatible with the alteration model presented by Young et al. (1999).

Tagish Lake is a unique C2 meteorite. It might be related to the Antarctic micrometeorites which also represent anomalous C2 matter. Tagish Lake probably gives us the first well preserved large sample of the C2 matter that dominates interplanetary matter since the formation of the solar system (see Gounelle et al., 2001a). The model proposed by Young et al. (1999) cannot however explain such a domination of C2 matter over the other types of carbonaceous chondrites matter.

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TABLE 1a : Oxygen isotopic composition of anhydrous (primary) minerals of Tagish Lake – Carbonate-poor lithology.

Analysis number	Mineral <sup>†</sup>	Mineral composition	$\delta^{18}\text{O} \pm 1\sigma$ (‰)	$\delta^{17}\text{O} \pm 1\sigma$ (‰)	$\Delta^{17}\text{O} \pm 1\sigma$ (‰)
TL2 #18	Ol	Fe <sub>38.0</sub>	7.4 ± 1.1	5.4 ± 1.4	1.4 ± 1.5
TL3_1 #11	Ol	Fe <sub>94.8</sub>	7.2 ± 1.3	4.7 ± 1.3	1.0 ± 1.2
TL2 #16	Ol	Fe <sub>95.2</sub>	4.8 ± 1.2	3.7 ± 1.2	1.2 ± 1.3
TL3_6 #2	Ol	Fe <sub>97.5</sub>	2.7 ± 1.3	-1.5 ± 1.2	-2.9 ± 1.0
TL3_2 #9	Ol	Fe <sub>99.1</sub>	2.6 ± 1.3	-1.9 ± 1.2	-3.2 ± 1.1
TL3_6 #4	Ol	Fe <sub>69.6</sub>	1.2 ± 1.4	0.3 ± 1.2	-0.3 ± 1.1
TL3_1 #9	Ol	Fe <sub>80.7</sub>	0.0 ± 1.2	-0.7 ± 1.2	-0.7 ± 1.1
TL3_1 #27	Ol	Fe <sub>94.8</sub>	-0.3 ± 1.3	-1.7 ± 1.1	-1.5 ± 0.9
TL3_3 #2	Ol	Fe <sub>55.7</sub>	-0.6 ± 1.1	-3.7 ± 1.3	-3.5 ± 1.1
TL3_2 #11	Ol	Fe <sub>99.0</sub>	-1.3 ± 1.3	-4.1 ± 1.3	-3.4 ± 1.2
TL3_2 #10	Ol	Fe <sub>99.0</sub>	-1.5 ± 1.4	-3.8 ± 1.4	-3.0 ± 1.2
TL3_2 #13	Ol	Fe <sub>80.0</sub>	-2.1 ± 1.4	-0.8 ± 1.3	0.3 ± 1.1
TL3_1 #3*	Ol	Fe <sub>98.3</sub>	-2.1 ± 1.2	-6.5 ± 1.2	-5.4 ± 1.0
TL3_5 #4	Ol	Fe <sub>99.2</sub>	-2.2 ± 1.4	-6.7 ± 1.4	-5.6 ± 1.3
TL3_6 #5	Ol	Fe <sub>97.0</sub>	-2.6 ± 1.7	-2.7 ± 1.5	-1.3 ± 1.5
TL2 #4	Ol	Fe <sub>99.2</sub>	-2.8 ± 1.3	-4.6 ± 1.3	-3.2 ± 1.4
TL2 #15	Ol	Fe <sub>98.9</sub>	-2.8 ± 1.6	-7.6 ± 1.4	-6.1 ± 1.6
TL3_2 #4	Ol	Fe <sub>99.1</sub>	-2.9 ± 1.3	-7.4 ± 1.4	-5.9 ± 1.2
TL2 #3	Ol	Fe <sub>99.3</sub>	-3.3 ± 1.3	-5.4 ± 1.3	-3.7 ± 1.5
TL3_1 #1*	Ol	Fe <sub>98.5</sub>	-3.4 ± 1.2	-7.1 ± 1.2	-5.3 ± 1.0
TL2 #1	Ol	Fe <sub>99.0</sub>	-3.7 ± 1.3	-5.6 ± 1.2	-3.7 ± 1.4
TL3_6 #1	Ol	Fe <sub>99.3</sub>	-3.7 ± 1.4	-6.5 ± 1.2	-4.5 ± 1.0
TL3_5b #1	Ol	Fe <sub>98.8</sub>	-4.1 ± 1.4	-8.1 ± 1.1	-6.0 ± 1.0
TL3_5 #3	Ol	Fe <sub>69.0</sub>	-4.2 ± 1.5	-5.6 ± 1.4	-3.4 ± 1.3
TL3_1 #26*	Ol	Fe <sub>98.5</sub>	-4.3 ± 1.3	-5.2 ± 1.2	-2.9 ± 1.1
TL3_1 #2*	Ol	Fe <sub>98.5</sub>	-4.5 ± 1.2	-6.8 ± 1.2	-4.5 ± 1.1
TL3_4 #12	Ol	Fe <sub>98.8</sub>	-4.6 ± 1.4	-5.9 ± 1.1	-3.5 ± 1.0
TL3_1 #10	Ol	Fe <sub>99.1</sub>	-4.8 ± 1.5	-8.1 ± 1.5	-5.6 ± 1.4
TL2 #17	Ol	Fe <sub>99.0</sub>	-4.8 ± 1.2	-8.7 ± 1.3	-6.1 ± 1.4
TL3_2 #12	Ol	Fe <sub>98.9</sub>	-5.3 ± 1.3	-7.8 ± 1.3	-5.0 ± 1.2
TL3_1 #7	Ol	Fe <sub>42.5</sub>	-5.4 ± 1.2	-4.5 ± 1.2	-1.7 ± 1.1
TL2 #19	Ol	Fe <sub>73.6</sub>	-5.4 ± 1.3	-7.5 ± 1.2	-4.7 ± 1.3
TL3_1 #8	Ol	Fe <sub>44.8</sub>	-5.6 ± 1.3	-5.1 ± 1.3	-2.2 ± 1.1
TL3_6 #3	Ol	Fe <sub>99.1</sub>	-6.0 ± 1.4	-9.6 ± 1.1	-6.5 ± 1.0
TL3_2 #3	Ol	Fe <sub>99.3</sub>	-6.7 ± 1.3	-9.2 ± 1.2	-5.6 ± 1.0
TL3_5 #2	Ol	Fe <sub>98.4</sub>	-7.6 ± 1.4	-8.9 ± 1.3	-5.0 ± 1.1
TL3_2 #7	Ol	Fe <sub>99.2</sub>	-7.6 ± 1.7	-8.6 ± 2.4	-4.7 ± 2.4
TL3_2 #2	Ol	Fe <sub>99.3</sub>	-7.8 ± 1.5	-8.4 ± 1.5	-4.3 ± 1.4
TL3_2 #5	Ol	Fe <sub>98.9</sub>	-9.2 ± 1.3	-14.1 ± 1.3	-9.3 ± 1.1
TL3_2 #1	Ol	Fe <sub>99.1</sub>	-10.1 ± 1.4	-13.4 ± 1.2	-8.1 ± 1.1
TL3_6 #6@1	Ol	Fe <sub>63.6</sub>	-10.5 ± 1.4	-10.5 ± 1.3	-5.0 ± 1.2
TL2 #14	Ol	Fe <sub>98.5</sub>	-40.1 ± 1.5	-42.3 ± 1.2	-21.4 ± 1.4
TL2 #13	Ol	Fe <sub>99.1</sub>	-41.0 ± 1.2	-40.2 ± 1.3	-18.8 ± 1.5
TL3_1 #14	Ol	Fe <sub>99.5</sub>	-46.1 ± 1.4	-47.9 ± 1.3	-24.0 ± 1.2
TL3_1 #13	Ol	Fe <sub>99.5</sub>	-46.2 ± 1.2	-48.7 ± 1.3	-24.6 ± 1.1
TL3_1 #15	Px	En <sub>54.5</sub> Wo <sub>1.2</sub>	8.8 ± 2.2	5.0 ± 2.5	0.5 ± 2.6
TL3_4 #13	Px	En <sub>92.0</sub> Wo <sub>2.8</sub>	-0.7 ± 1.6	-5.8 ± 1.1	-5.5 ± 1.1
TL3_2 #14	Px	En <sub>96.2</sub> Wo <sub>2.4</sub>	-10.0 ± 1.5	-11.8 ± 1.3	-6.6 ± 1.2

\*The analyses are ordered by decreasing  $\delta^{18}\text{O}$  for the anhydrous minerals, and by decreasing  $\Delta^{17}\text{O}$  for the secondary minerals (carbonates, magnetite, matrix).

<sup>†</sup>The following abbreviations are used : Ol = olivine, Px = pyroxene, Mes. = mesostatis, Carb. = carbonate, Mx = matrix, Mt = magnetite.

\*LIME olivines (MnO = 1.3wt%)

TABLE 1b : Oxygen isotopic composition of secondary minerals of Tagish Lake – Carbonate-poor lithology.

Analysis number	Mineral <sup>†</sup>	Mineral composition	$\delta^{18}\text{O} \pm 1\sigma$ (‰)	$\delta^{17}\text{O} \pm 1\sigma$ (‰)	$\Delta^{17}\text{O}$ (‰)
TL3_3 #4 <sup>‡</sup>	Carb. + Mx	Mg* = 63.6	19.5 ± 1.1	12.7 ± 1.3	2.5 ± 2.1
TL3_1 #33	Carb. + Mx	Mg* = 49.9	25.5 ± 1.6	14.0 ± 1.5	0.7 ± 1.9
TL3_1 #24	Mt	Fe <sub>3</sub> O <sub>4</sub>	-8.9 ± 1.4	-0.2 ± 0.9	4.4 ± 0.9
TL3_3 #1	Mt	Fe <sub>3</sub> O <sub>4</sub>	-7.9 ± 1.7	-0.4 ± 1.1	3.7 ± 1.2
TL3_1 #23	Mt	Fe <sub>3</sub> O <sub>4</sub>	-6.2 ± 1.8	0.0 ± 1.2	3.2 ± 1.3
TL3_1 #28	Mt	Fe <sub>3</sub> O <sub>4</sub>	-9.0 ± 1.5	-2.2 ± 1.4	2.5 ± 1.3
TL3_1 #29	Mt + Mx	n.a.	-5.8 ± 1.9	-1.9 ± 1.5	1.1 ± 1.6
TL3_1 #18	Mt + Mx	Mg* = 55.5	-7.6 ± 1.9	-2.9 ± 1.6	1.0 ± 1.6
TL3_1 #22	Mt + Mx	Mg* = 0.6	-5.5 ± 1.4	-2.2 ± 1.2	0.6 ± 1.2
TL3_5 #5	Mt + Mx	Mg* = 0.3	-6.2 ± 1.4	-2.8 ± 1.6	0.4 ± 1.4
TL2 #20	Mx	Mg* = 5.5	11.2 ± 1.3	9.0 ± 1.2	3.1 ± 1.4
TL2 #2	Mx	Mg* = 59.5	16.3 ± 1.4	11.4 ± 1.3	2.9 ± 1.4
TL2 #8	Mx	Mg* = 34.3	9.3 ± 1.6	7.4 ± 1.3	2.6 ± 1.5
TL3_1 #32	Mx	Mg* = 49.9	12.0 ± 1.9	8.4 ± 1.6	2.1 ± 1.7
TL2 #10	Mx	Mg* = 46.5	4.6 ± 1.2	4.1 ± 1.3	1.7 ± 1.4
TL2 #11	Mx	n.a.	11.7 ± 1.9	7.7 ± 1.9	1.6 ± 2.2
TL3_4 #14	Mx	Mg* = 60.9	12.1 ± 1.3	7.7 ± 1.3	1.4 ± 1.1
TL3_1 #17	Mx	Mg* = 63.4	11.8 ± 1.9	6.8 ± 2.0	0.7 ± 2.0
TL3_1 #30	Mx	Mg* = 70.9	19.7 ± 1.3	10.2 ± 1.2	-0.1 ± 1.1
TL2 #9	Mx	Mg* = 53.4	14.0 ± 1.5	7.1 ± 1.5	-0.2 ± 1.4
TL2 #5	Mx	n.a.	8.3 ± 1.6	4.2 ± 1.1	-0.2 ± 1.7
TL2 #12	Mx	n.a.	16.1 ± 1.9	8.1 ± 1.8	-0.3 ± 2.0
TL3_1 #16	Mx	Mg* = 78.4	16.0 ± 1.4	8.0 ± 1.3	-0.3 ± 2.3
TL2 #7	Mx	Mg* = 42.0	14.1 ± 2.6	7.0 ± 1.9	-0.3 ± 1.2
TL2 #6	Mx	Mg* = 43.5	6.1 ± 1.6	2.5 ± 1.4	-0.7 ± 1.6
TL3_5 #1	Mx	Mg* = 70.4	9.8 ± 2.0	4.1 ± 1.5	-1.0 ± 1.5
TL3_3 #3	Mx	Mg* = 61.8	6.6 ± 1.0	1.7 ± 2.5	-1.7 ± 2.4
TL3_1 #19	Mx	Mg* = 55.6	3.6 ± 1.8	0.1 ± 1.4	-1.8 ± 1.4

<sup>‡</sup> The analyses are ordered by decreasing  $\delta^{18}\text{O}$  for the anhydrous minerals, and by decreasing  $\Delta^{17}\text{O}$  for the secondary minerals (carbonates, magnetite, matrix).

<sup>†</sup> The following abbreviations are used : Ol = olivine, Px = pyroxene, Mes. = mesostatis, Carb. = carbonate, Mx = matrix, Mt = magnetite n.a. : not analyzed.

<sup>‡</sup> weighted mean of two analyses in the same spot.

TABLE 2 : Oxygen isotopic composition of minerals in Tagish Lake\* – Carbonate-rich lithology.

Analysis number	Mineral †	Mineral composition	$\delta^{18}\text{O} \pm 1\sigma$ (‰)	$\delta^{17}\text{O} \pm 1\sigma$ (‰)	$\Delta^{17}\text{O} \pm 1\sigma$ (‰)
TL1 #32	Ol	Fe <sub>98.7</sub>	4.1 ± 0.8	0.3 ± 1.0	-1.8 ± 1.2
TL1 #14	Ol	Fe <sub>99.0</sub>	3.1 ± 0.9	-1.8 ± 0.7	-3.5 ± 0.8
TL1 #29	Ol	Fe <sub>99.2</sub>	-1.2 ± 0.6	-5.6 ± 0.8	-4.9 ± 1.0
TL1 #30	Ol	Fe <sub>98.6</sub>	-2.3 ± 0.7	-5.0 ± 1.0	-3.8 ± 1.2
TL1 #13	Ol	Fe <sub>98.7</sub>	-2.5 ± 1.1	-6.6 ± 0.7	-5.3 ± 0.8
TL1 #17	Ol	Fe <sub>91.3</sub>	-2.8 ± 1.0	-5.2 ± 0.5	-3.8 ± 0.6
TL1 #31	Ol	Fe <sub>98.6</sub>	-3.5 ± 0.9	-6.6 ± 0.8	-4.8 ± 1.1
TL1 #16	Ol	Fe <sub>98.6</sub>	-3.9 ± 0.9	-7.7 ± 0.7	-5.7 ± 0.7
TL1 #15	Ol	Fe <sub>98.8</sub>	-4.8 ± 1.0	-8.3 ± 0.5	-5.8 ± 0.7
TL1 #21	Ol	Fe <sub>99.2</sub>	-5.2 ± 0.9	-7.7 ± 0.7	-5.0 ± 0.7
TL1 #23	Ol	Fe <sub>99.2</sub>	-7.1 ± 1.0	-10.7 ± 0.6	-7.0 ± 0.7
TL1 #26	Ol	Fe <sub>99.4</sub>	-40.6 ± 1.0	-43.4 ± 0.7	-22.3 ± 1.0
TL1 #25	Ol	Fe <sub>99.4</sub>	-42.5 ± 0.7	-43.3 ± 0.7	-21.3 ± 0.9
TL1 #6	Px + Mes.	En <sub>94.5</sub>	8.4 ± 1.1	2.1 ± 0.7	-2.3 ± 0.8
TL1 #8	Px + Mes.	En <sub>91.1</sub> Wo <sub>5.2</sub>	6.4 ± 0.9	2.7 ± 0.6	-0.6 ± 0.7
TL1 #1	Px	En <sub>93.0</sub> Wo <sub>3.8</sub>	-6.2 ± 1.0	-7.3 ± 0.6	-4.1 ± 0.7
TL1 #3	Px	En <sub>96.0</sub> Wo <sub>2.3</sub>	-6.4 ± 1.0	-8.0 ± 0.7	-4.7 ± 0.8
TL1 #4	Px	En <sub>97.6</sub> Wo <sub>1.1</sub>	-6.5 ± 0.9	-7.6 ± 0.6	-4.3 ± 0.7
TL1 #2	Px	En <sub>96.4</sub> Wo <sub>1.8</sub>	-8.7 ± 0.9	-9.4 ± 0.6	-4.9 ± 0.6
TL1 #24	Carb.	CaCO <sub>3</sub>	25.0 ± 0.8	13.7 ± 0.5	0.7 ± 0.7
TL1 #12	Carb.	CaCO <sub>3</sub>	35.3 ± 1.4	18.8 ± 1.2	0.5 ± 0.5
TL1 #11	Mx	Mg* = 42.7	1.4 ± 1.9	3.6 ± 1.8	2.9 ± 2.9
TL1 #20	Mx	Mg* = 49.1	5.8 ± 1.2	4.3 ± 0.9	1.3 ± 1.3
TL1 #27	Mx	Mg* = 59.8	12.3 ± 0.6	7.5 ± 0.8	1.0 ± 1.0
TL1 #19	Mx	Mg* = 47.0	17.8 ± 1.4	9.9 ± 0.9	0.6 ± 0.6
TL1 #18	Mx	Mg* = 47.3	20.3 ± 1.3	9.8 ± 1.0	-0.8 ± -0.8
TL1 #9	Mx	Mg* = 57.3	7.8 ± 0.9	3.3 ± 0.8	-0.8 ± -0.8
TL1 #10	Mx	Mg* = 51.8	14.5 ± 1.1	6.4 ± 0.8	-1.1 ± -1.1
TL1 #22	Mx	Mg* = 83.0	11.8 ± 1.7	4.7 ± 0.8	-1.4 ± -1.4
TL1 #28	Mx	Mg* = 60.4	7.5 ± 1.6	2.0 ± 1.1	-1.8 ± -1.8

\*The analyses are ordered by decreasing  $\delta^{18}\text{O}$  values for the anhydrous minerals, and by decreasing  $\Delta^{17}\text{O}$  for the secondary minerals (carbonates, magnetite, matrix).

†The following abbreviations are used : Ol = olivine, Px = pyroxene, Mes. = mesostatis, Carb. = carbonate, Mx = matrix.



## Figure captions

**Figure 1** : Electron micrograph mosaic (backscattered images) of a fragment of the dominant lithology of Tagish lake, the carbonate-poor lithology. See (Zolensky et al., 2001) for a detailed description of this lithology. Note the bright spots which consist mainly of magnetite in the framboidal form.

**Figure 2** : Backscattered electron micrograph of the Tagish Lake matrix, carbonate-rich lithology (see text or Zolensky et al., 2001 for details about its mineralogy).

**Figure 3** : Three isotope diagram representing the oxygen isotopic composition of the meteorite Tagish Lake (carbonate-poor lithology) measured by ion microprobe (Cameca IMS1270, Nancy). The terrestrial fractionation line (TF), the line defined by the analyses of Allende CAIs (CAI) and the Young et Russell (Y&R) are indicated for reference.

**Figure 4** : Three isotope diagram representing the oxygen isotopic composition of the meteorite Tagish Lake (carbonate-rich lithology) measured by ion microprobe (Cameca IMS1270, Nancy). The terrestrial fractionation line (TF), the line defined by the analyses of Allende CAIs (CAI) and the Young et Russell (Y&R) are indicated for reference.

**Figure 5** : Three isotope diagram representing the oxygen isotopic composition of primary minerals from both lithologies measured by ion microprobe. The terrestrial

fractionation line (TF), the line defined by the analyses of Allende CAIs (CAI) and the Young et Russell (Y&R) are indicated for reference.

**Figure 6 :** Three isotope diagram representing the oxygen isotopic composition of secondary minerals (matrix, carbonate, and magnetite grains) from both lithologies measured by ion microprobe. The bulk oxygen isotopic composition of Tagish Lake (Brown et al., 2000) and the carbonate analyses from Leshin et al. (2001) are represented for comparison purposes. The terrestrial fractionation line (TF), the line defined by the analyses of Allende CAIs (CAI) and the Young et Russell (Y&R) are indicated for reference.

**Figure 7 :** Electron micrograph of one  $^{16}\text{O}$ -rich olivine inclusion.

**Figure 8 :** Oxygen isotopic composition of Tagish Lake olivine minerals as a function of their FeO content. This trend is similar to what is found in other carbonaceous chondrites and in micrometeorites (see Leshin et al., 1997; Engrand et al., 1999b; Leshin et al., 2000). Figure 5b corresponds to an enlargement of the iron-poor region of the graph.

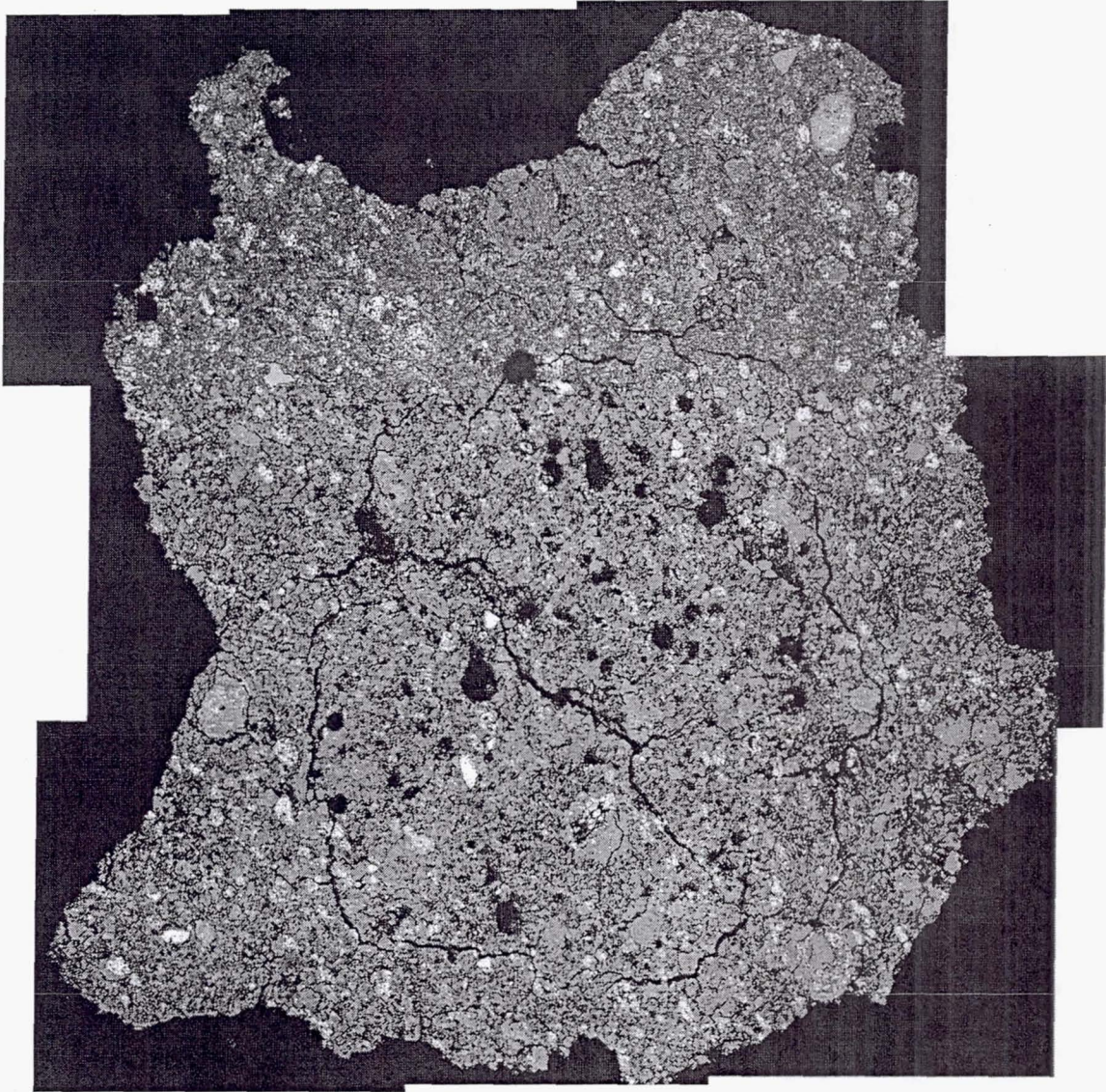
**Figure 9 :** Comparison of the oxygen isotopic composition of Tagish Lake matrix with carbonaceous chondrite matrix/whole rock (WR) data. Carbonaceous chondrite data is taken from Rowe *et al.* (1994); Clayton et al. (1976; 1977), Clayton and Mayeda (1984; 1999); Mayeda et al. (1987).

**Figure 10 :** Comparison of the oxygen isotopic composition of Tagish Lake carbonates with Orgueil (Leshin et al., 2001) and CM2 chondrites (Benedix et al., 2000). Additional Tagish Lake carbonate data are taken from (Leshin et al., 2001).

**Figure 11 :** Three isotope diagram representing the oxygen isotopic composition of magnetite grains of the carbonate-poor lithology measured by ion microprobe. Data for CI1 chondrites (Rowe et al., 1994), Essebi (CM2, Rowe et al., 1994), Renazzo (CR2, Weisberg et al., 1993), R-chondrites (Greenwood et al., 2000), and LL3 ordinary chondrites (Choi et al., 1998) are represented for comparison. Data were obtained by conventional analyses of mineral separates (ms), or by *in-situ* ion microprobe analyses (SIMS).



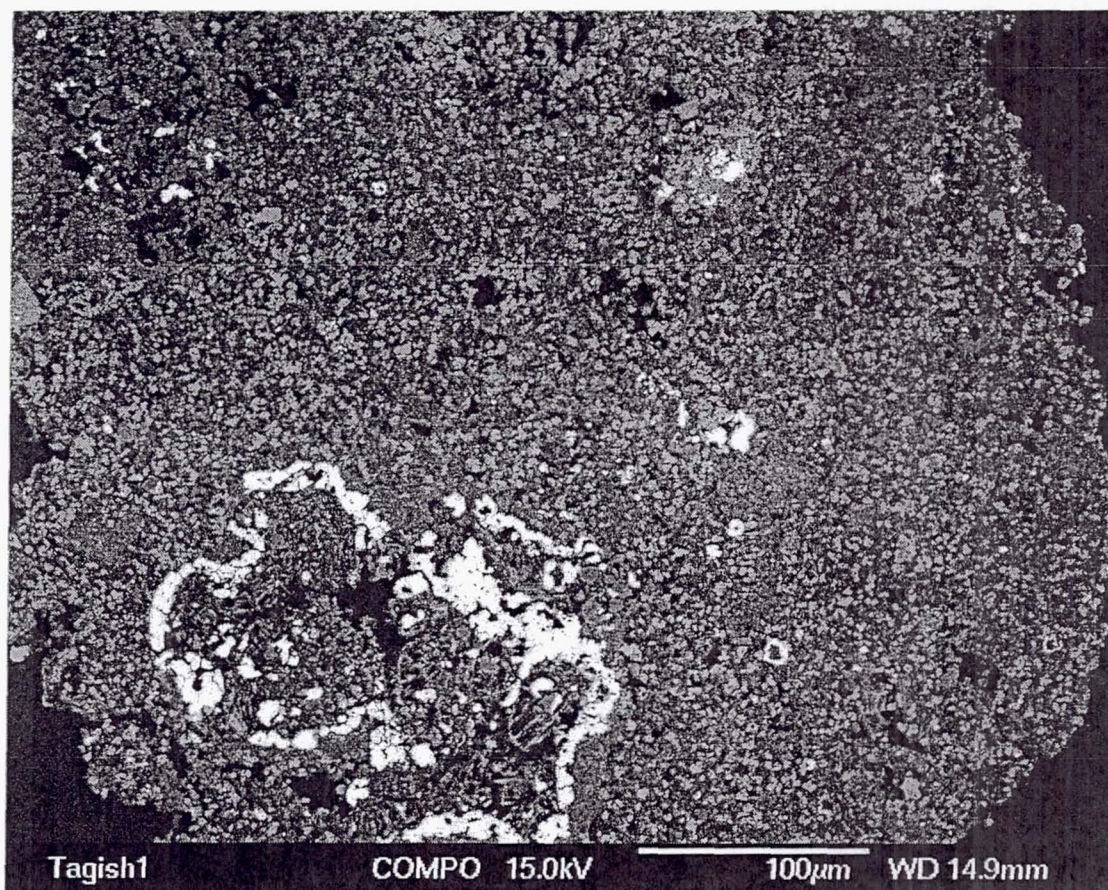
Figure 1



Engrand et al, 2001



Figure 2



Engrand et al, 2001

Figure 3

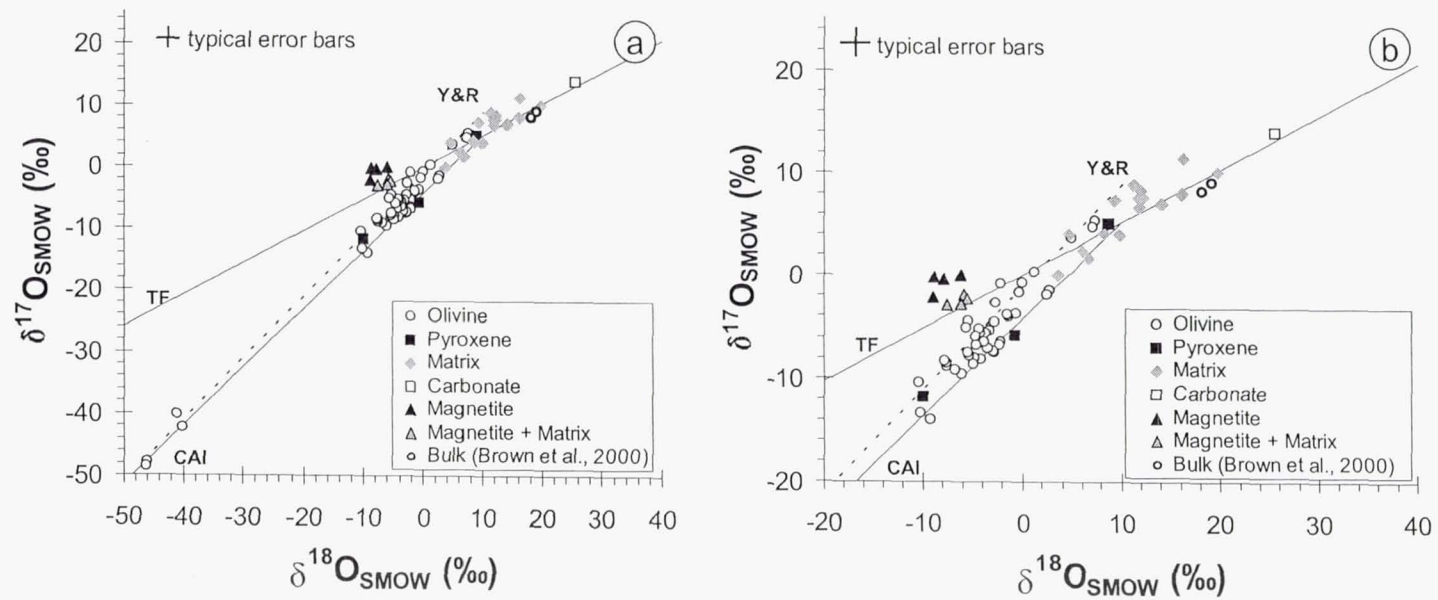
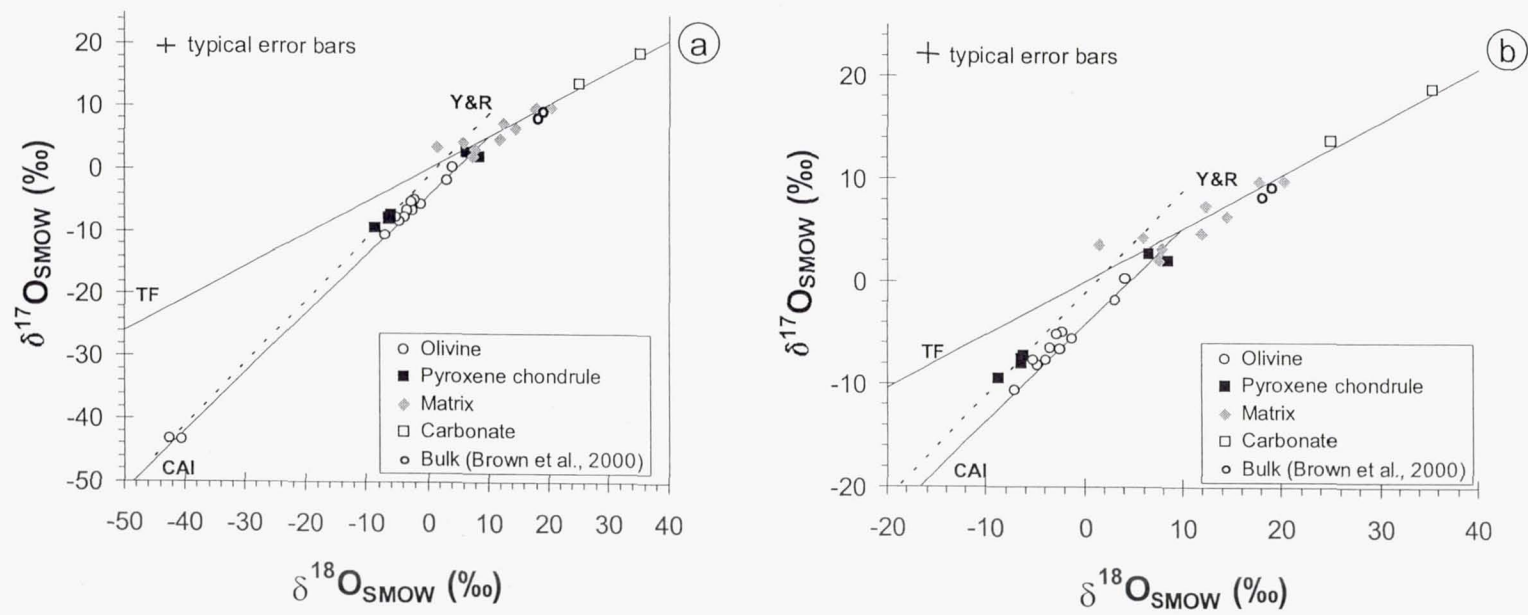
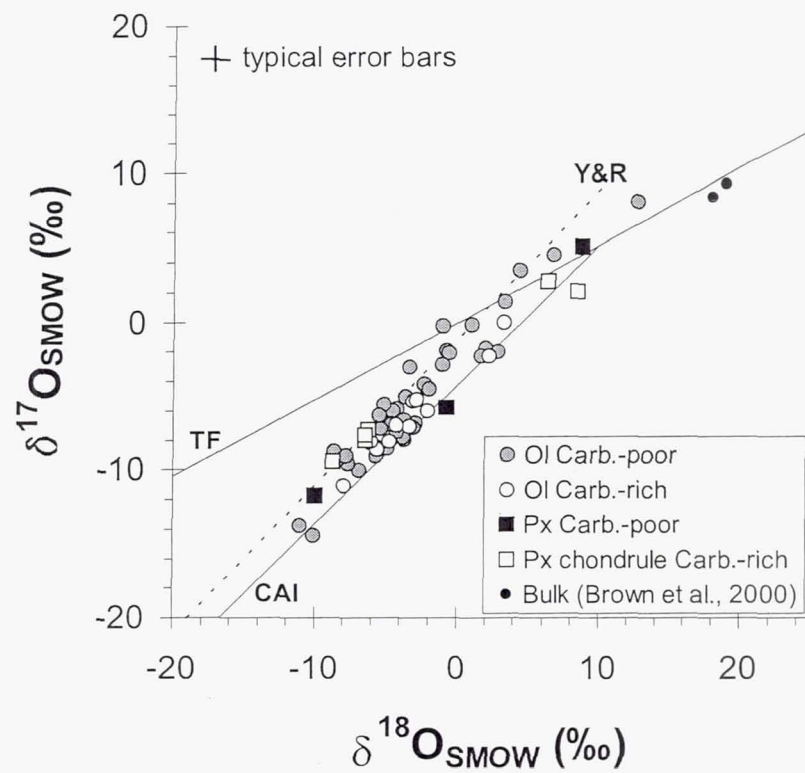


Figure 4



Engrand et al., 2001

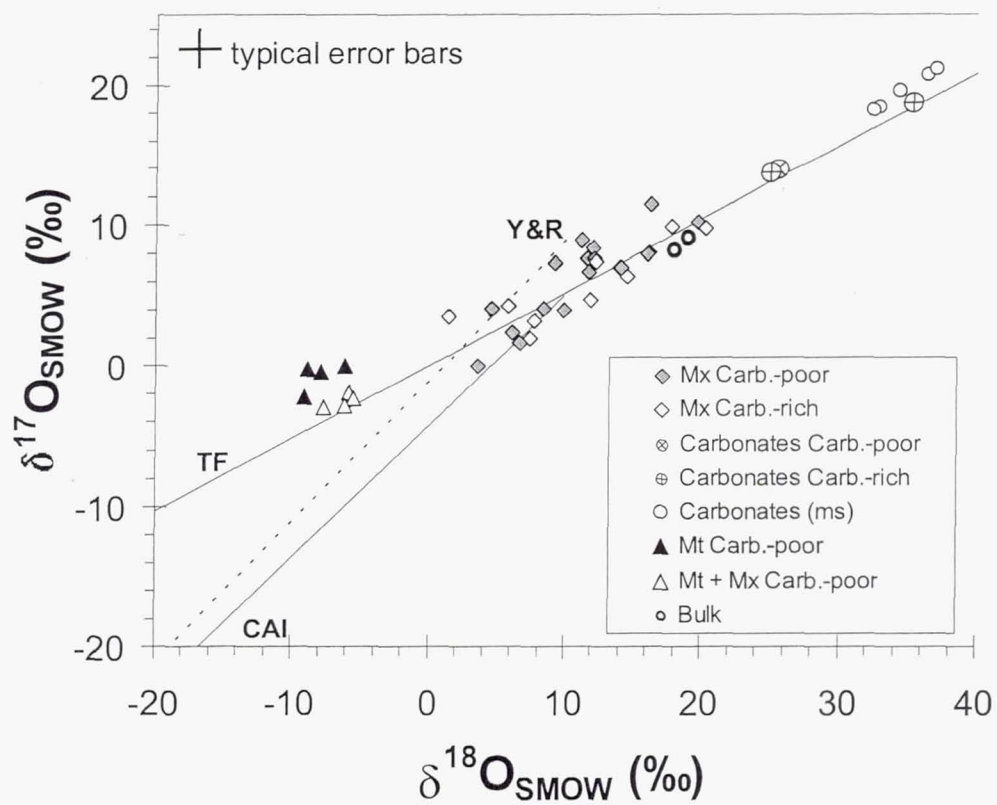
Figure 5



Engrand et al., 2001

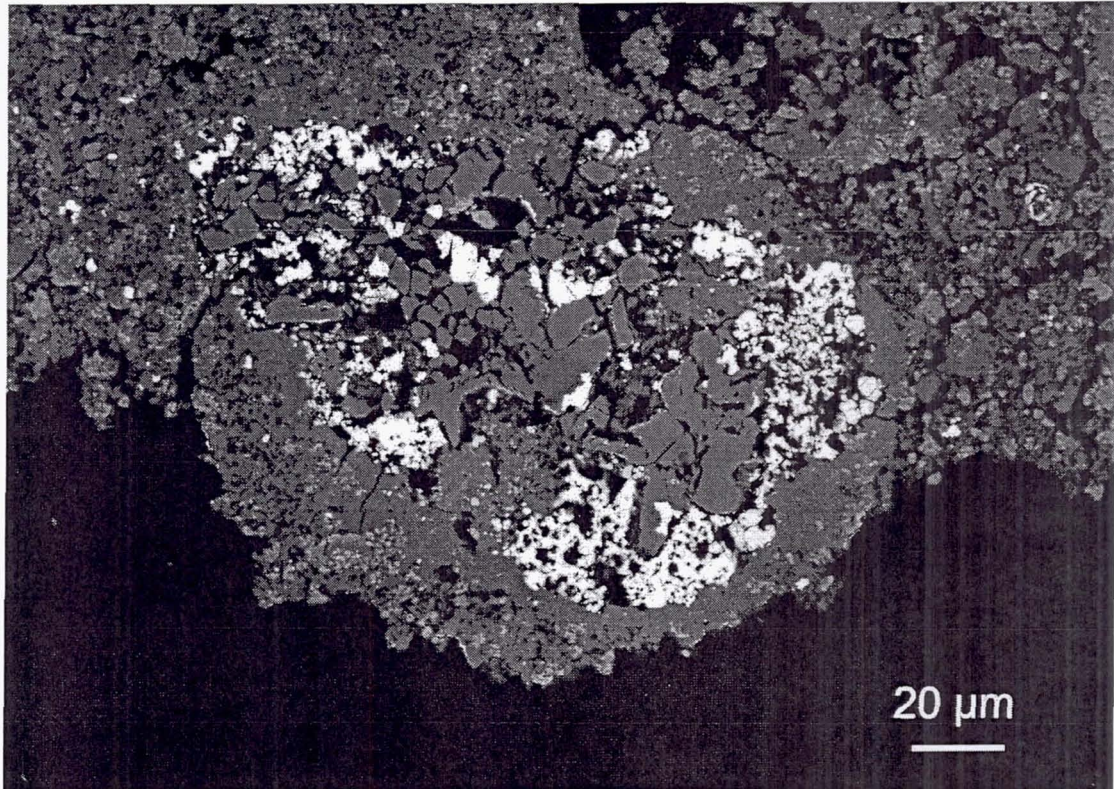


Figure 6



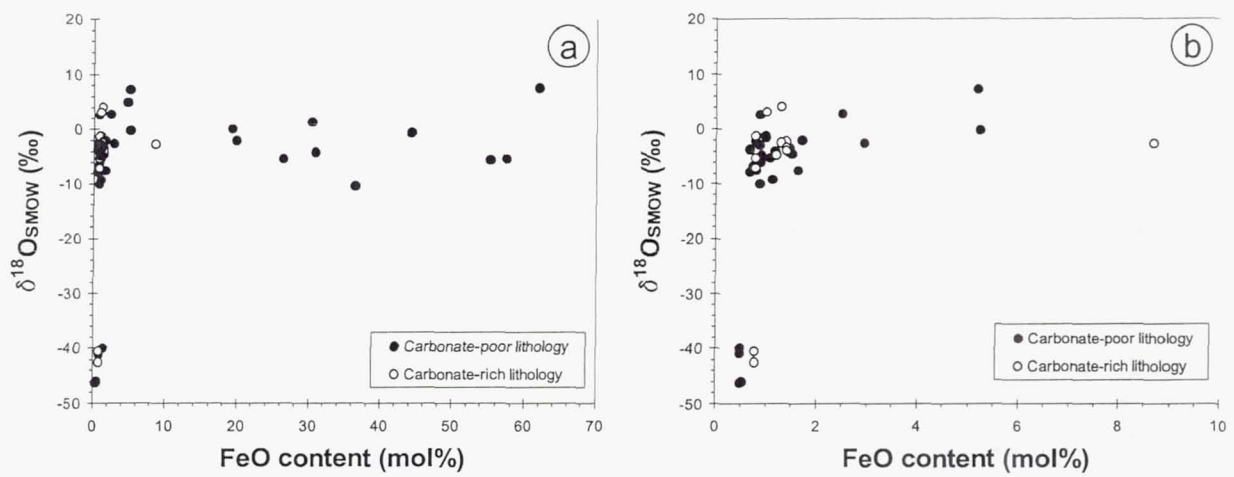
Engrand et al., 2001

Figure 7



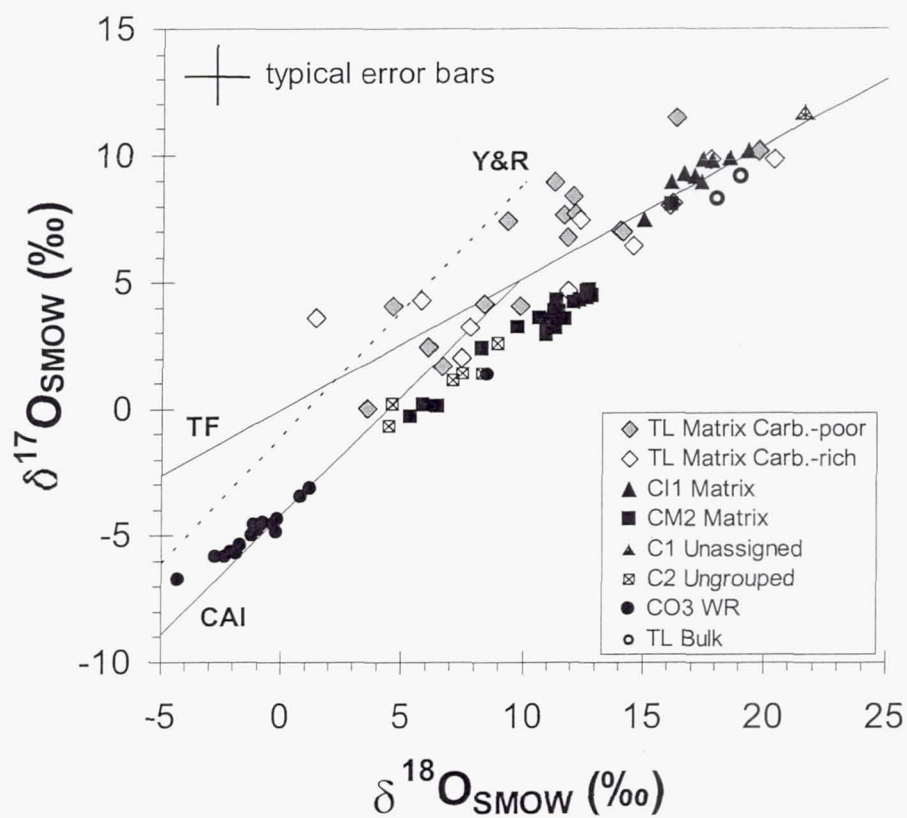
Engrand et al., 2001

Figure 8



Engrand et al., 2001

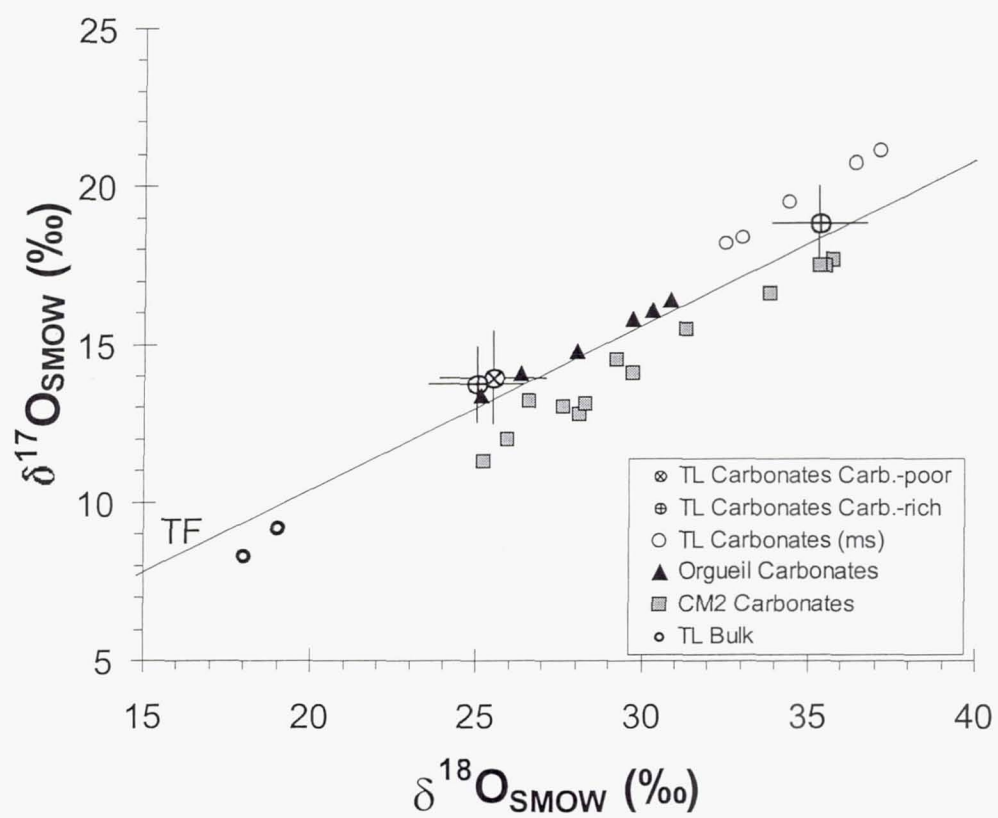
Figure 9



Engrand et al., 2001

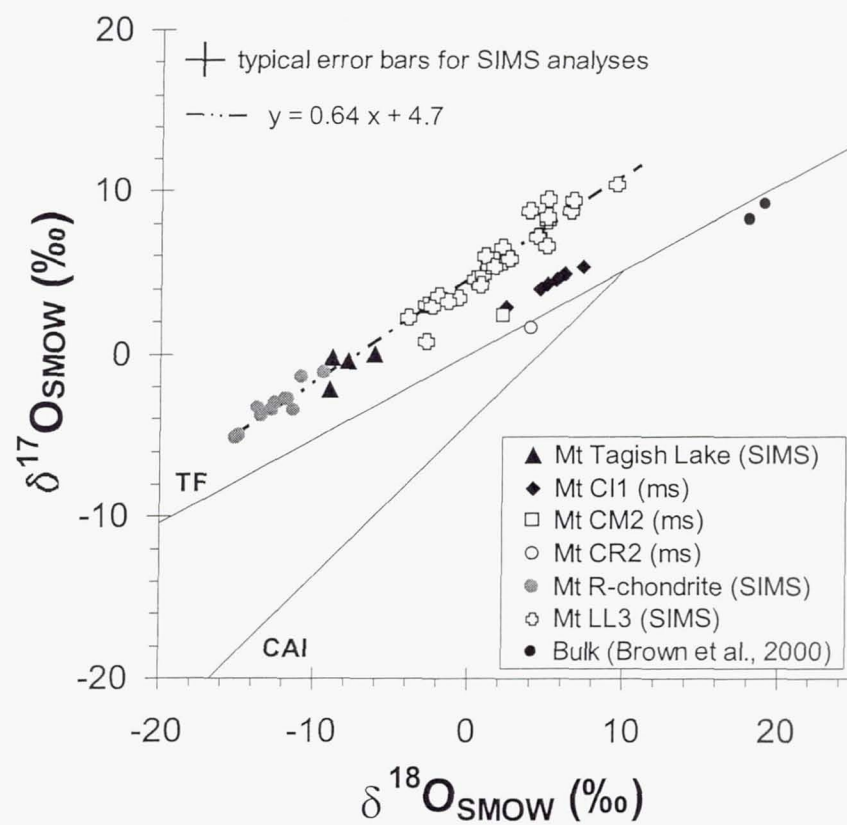


Figure 10



Engrand et al., 2001

Figure 11



Engrand et al., 2001